7. Crystal Activation

Experiment MA-151

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ABSTRACT

The Crystal Activation Experiment consisted of two sample packages that were flown in the command module and returned to Earth for analysis of the radioactivity induced in them during the flight. The objective of the experiment was to define the background caused by detector activation that interferes when gamma radiation is measured in the 0.02- to 10-MeV range from Earth orbit. These measurements, together with accelerator beam activation measurements and theoretical calculations, will be used to estimate this background for future or planned experiments. To aid the calculations, metals selected for their cross sections for specific neutron and proton energies were included in the samples in addition to the germanium and thallium-activated sodium iodide (NaI(Tl)), which are the usual detectors for low-energy gamma-ray measurements.

Preliminary results show that the activation of the NaI(Tl) crystal was a factor of 3 below that from a similar measurement on Apollo 17. The identification of certain species and the level of activation observed show an important contribution from the interactions of thermal and energetic neutrons produced as secondaries in the spacecraft. The reduction of the activation by a factor of 3, compared with the Apollo 17 experiment, is consistent with the reduction in the primary cosmic ray intensity due to the geomagnetically shielded orbit.

INTRODUCTION

Significant background components to X-ray and gamma-ray measurements performed in space can be attributed to X-ray and gamma-ray emission induced in the spacecraft surrounding the detector and within the detector itself. Varying detector-spacecraft geometries (such as those obtainable with an extendable boom) can provide an indication of the spacecraft contribution, but in-flight measurements cannot separate the cosmic-ray-induced detector background from counts caused by the external photon flux.

Previous measurements were made on an NaI(Tl) crystal flown on Apollo 17. For that flight, the activation was mainly induced by cosmic-ray protons and by secondary neutrons produced in the spacecraft material. Because of the low-altitude Earth orbit on the Apollo-Soyuz Test Project (ASTP) mission, the Apollo-Soyuz spacecraft was exposed to a proton flux that was less intense. However, the protons had a higher

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average energy and therefore could have been more efficient in producing secondary neutrons. Neither the particle flux inside the spacecraft nor the cross sections for production of radioactive isotopes in the detector materials are well known. Therefore, in addition to NaI(Tl) and germanium detector crystals, the sample package included a number of metals that have good cross sections for specific energy ranges of neutrons and protons.

SCIENTIFIC BACKGROUND

One of the major difficulties in measuring 0.02- to 10-MeV gamma radiation from space lies in distinguishing the radiation emitted by cosmic and planetary sources from the high rate of background gamma radiation produced by cosmic-ray and trapped-proton interaction with the spacecraft and the detector materials. Of these effects, the most difficult to isolate is the delayed decay of a radioactive nucleus produced by a spallation interaction within the detector material. These effects particularly complicate measurements of the diffuse sky-background gamma radiation in a spectral band that could contain important information on cosmology and the role of antimatter in the universe. The experimental confusion and theoretical possibilities have been recently reviewed by Horstman, Cavallo, and Moretti-Horstman (ref. 7-1). It is evident that advances in the field require improved knowledge of local activation effects and optimization of detector design and location against such a background.

Radioactive background has been observed in all cosmic gamma-ray experiments that have been carried into regions of trapped radiation such as in the Orbiting Solar Observatory (OSO) series of spacecraft. Such effects have been successfully explained (ref. 7-2) by a model proposed by Dyer and Morfill (ref. 7-3) in which the direct proton interactions in scintillators lead to radioactive spalled nuclei, the decay products of which are indistinguishable from an external gamma ray. The work by Dyer and Morfill and that by Fishman (ref. 7-4) demonstrated that similar interactions due to cosmic rays could also cause contamination, and this has been verified by the gamma-ray NaI(Tl) spectrometers carried onboard Apollo 15 and Apollo 16 (ref. 7-5). As a result, an identical crystal was flown and returned on Apollo 17. Application of the spallation-activation calculation to those results showed that the level of activation was considerably enhanced by significant fluxes of secondary neutrons produced in the heavy spacecraft by cosmic-ray proton interactions (ref. 7-6). Results from activation packages flown on Skylab (ref. 7-7) also showed the presence of secondary neutrons but indicated less thermalization of this flux than that indicated by the Apollo 17 data.

For these reasons, it was considered important to include an activation package onboard the Apollo-Soyuz spacecraft to obtain further data from near-Earth orbit. For the Apollo-Soyuz orbit (222 km altitude, 51.8° inclination), part of the primary cosmic ray flux is shielded by the Earth's magnetic field, and trapped proton effects are minimized by the low orbit.

EQUIPMENT AND EXPERIMENTAL PROCEDURE

The two assemblies shown in figures 7-1 and 7-2 were flown in the command module of the Apollo spacecraft during the Apollo-Soyuz mission. An NaI(Tl) crystal (fig. 7-1) physically identical to those flown on the Apollo 15 and 16 missions was used in this experiment. This assembly did not include the photomultiplier, the proton anticoincidence mantle, and the thermal shield that were used during the Apollo 15 and 16 missions, but it was identical to the assembly used on the Apollo 17 mission. Thus, direct comparison of the results from the two missions can be made. The detector was a 7- by 7-cm right-cylindrical crystal, hermetically sealed in a steel cylinder with a glass plate at one end. Magnesium oxide was used as an optical reflector inside the crystal assembly except at the glass window. This assembly made possible the simple procedure for optically coupling the crystal to a photomultiplier tube after flight. The second assembly (fig. 7-2) consisted of a 724-g sample of high-purity germanium; a small intrinsic germanium detector 32 mm in diameter by 8.75 mm thick, with a 0.12-mm layer of indium

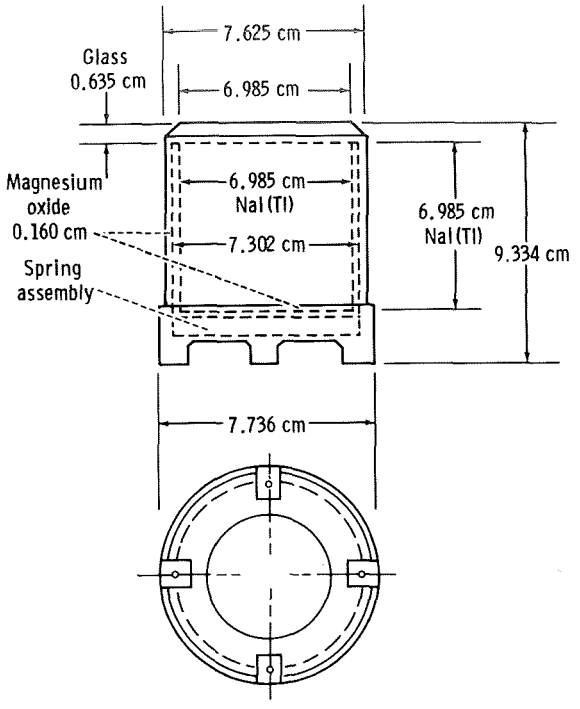

FIGURE 7-1.—The Apollo NaI(Tl) crystal assembly.

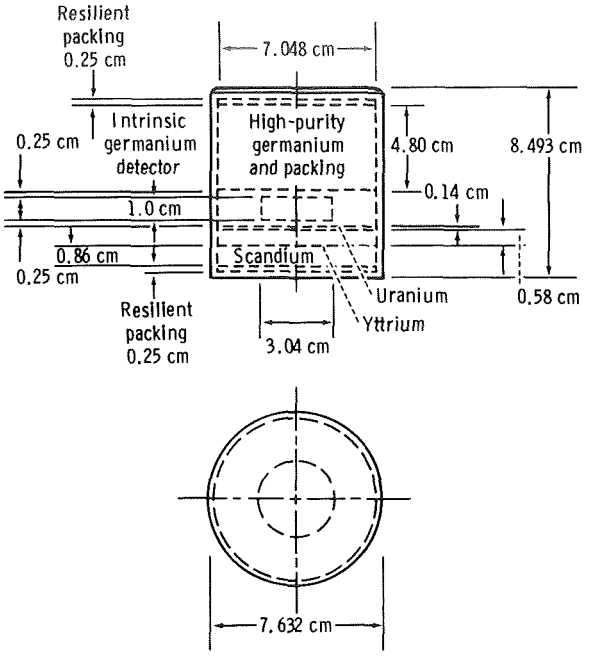

FIGURE 7-2.—Sample package showing location of metals and intrinsic germanium detector.

on one face; and 100-g foil disks of yttrium (Y), scandium (Sc), and depleted uranium (U) sealed in a cylinder identical with that used for containing the NaI(Tl) crystal. The lid of the container in this second assembly was made of 55 g of titanium (Ti) rather than steel.

Before the mission, background counts were taken on all materials in the laboratories where postflight low-level counting was anticipated. After splashdown, the flight (i.e., activated) NaI(Tl) was returned to the recovery ship and optically mounted on a photomultiplier tube, and pulse height spectra were obtained. The crystal counting was started approximately 1.5 hours after atmospheric entry of the command module. Before splashdown, a control (unactivated) crystal was optically sealed to the same photomultiplier tube, and the background was determined in the steel shield.

A 10.2- by 10.2-cm NaI(Tl) detector was used to measure the gamma-ray emission flux from the NaI(Tl) crystal and from the germanium after removal from its container. These measurements were also performed inside the steel shield, starting approximately 1.5 hours after atmospheric entry of the command module. Counting continued for 4 hours onboard the recovery carrier.

The detectors, bulk germanium, and foil disks were then returned to laboratories in the United States for further counting. The intrinsic germanium detector was returned to the Lawrence Berkeley Laboratory where the detector material was mounted on a cryostat, cooled, and low-level-counted. The foil disks and the steel container were delivered to the Los Alamos Scientific Laboratory for analysis. The NaI(Tl) crystals and the bulk germanium material were taken to the Oak Ridge National Laboratory, where the internal NaI(Tl) measurements were continued, and lithium-activated germanium (Ge(Li)) and low-level coincidence spectra were accumulated on both materials. High-resolution Ge(Li) spectra were also taken of the bulk germanium at the NASA Lyndon B. Johnson Space Center (JSC).

RESULTS

Recovery Ship Measurements

The internal NaI(Tl) spectrum taken onboard the recovery ship approximately 4 hours after reentry is compared with the equivalent data from the Apollo 17 experiment in figure 7-3. The radionuclides identified from the Apollo 17 data are shown. Although the statistics are poorer, the Apollo-Soyuz data have the same general shape and are lower in intensity by a factor of approximately 3. The bulk germanium data did not show any identifiable lines but indicated a low-energy excess consistent with the presence of gallium-67 (^{67}Ga) identified in the data taken at the Lawrence Berkeley Laboratory.

Lawrence Berkeley Laboratory

The detector was successfully brought into operation on the first attempt and showed the same noise and resolution characteristics that had been observed during preflight tests. Counting commenced at the Lawrence Berkeley Laboratory approximately 24 hours after splashdown.

The earliest spectra showed some excess count rates in continuum regions up to at least 1000 keV, and perhaps as high as 2000 keV; however, counting statistics will severely limit the quantitative results that can be obtained from these spectral distributions. Simple hand analysis indicated decay with (probably) more than one half-life, in the few-tens-of-hours range, but definitely shorter than the 78-hour half-life of ^{67}Ga.

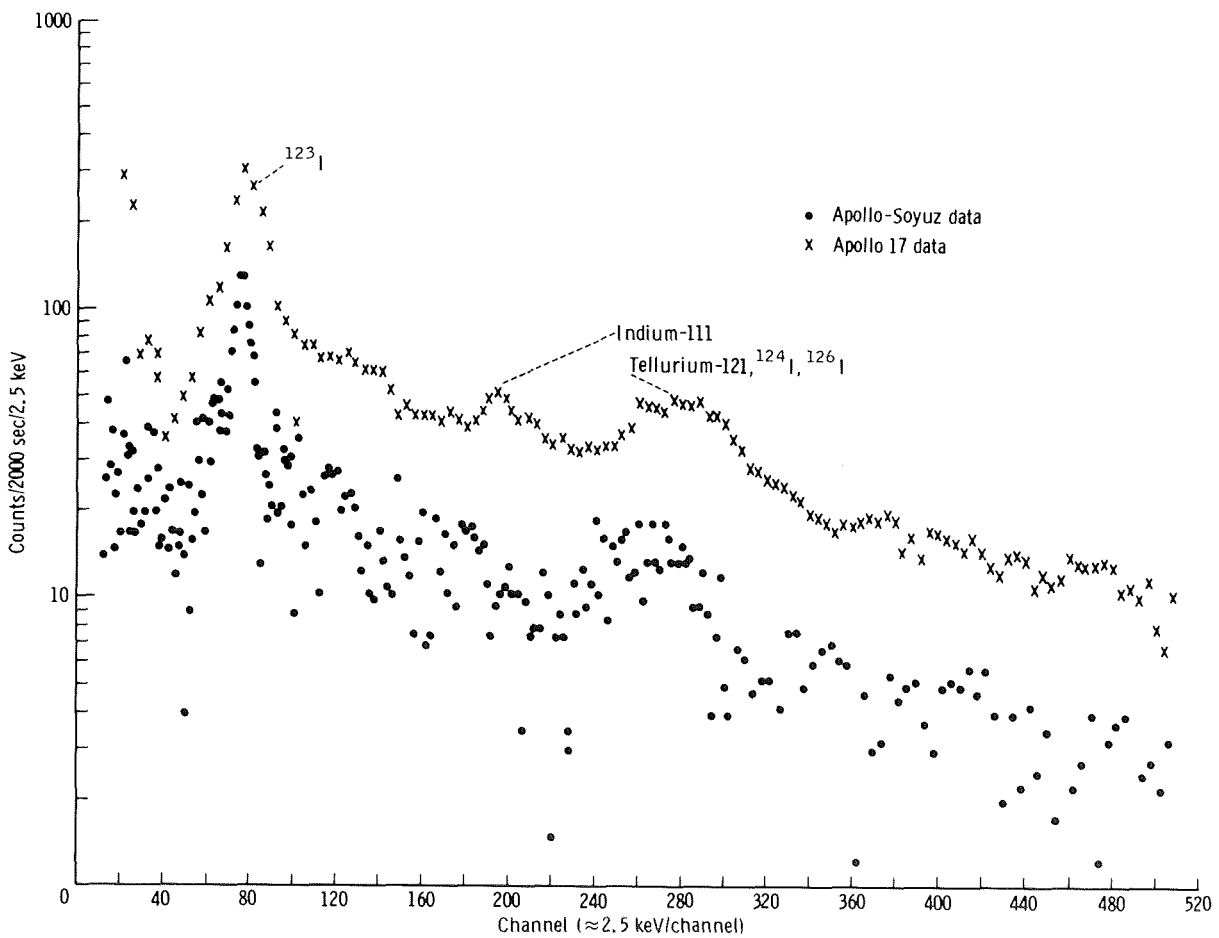

FIGURE 7-3.—Comparison of Apollo 17 and Apollo-Soyuz NaI(Tl) spectra taken 4 hours after reentry.

The prominent X-ray peak at approximately 10 keV (fig. 7-4) shows a composite decay curve that has been graphically resolved into two components, appropriate to 78-hour ^{67}Ga and 12-day germanium-71 (^{71}Ge). Identification of ^{67}Ga is conclusive, based on the aforementioned evidence, the decay of a 93-keV gamma ray with a single half-life of about 80 hours, the decay of a gamma ray with apparent energy of about 195 keV (the sum of the 185 keV gamma ray and the X-ray) having approximately the same half-life, and the delayed-coincidence results described in a later paragraph. Identification of ^{71}Ge is also considered conclusive, although it is based on less evidence, namely observation of the appropriate energy X-ray peak, which decays with (nearly) the ^{71}Ge half-life.

Identification of the 78-hour half-life radionuclide ^{67}Ga was confirmed from early spectra using delayed-coincidence measurements. In ^{67}Ga decay, the lowest level (93 keV) in the stable daughter zinc-67 (^{67}Zn) has a half-life of 9.3 microseconds; thus, the gamma ray that deexcites this level is almost always delayed in time with respect to emission of zinc fluorescent X-rays. Delayed-coincidence analysis was performed, using the X-ray events to generate a gate delayed 9 microseconds and of 27-microsecond width. A delayed-coincidence spectrum acquired during 2175 minutes of data accumulation that started 4 days after splashdown shows only the 93-keV peak and a scattering of lower energy partial absorption events. No events were stored in any of the 1420 channels above the peak region. This spectrum cannot be reconciled with any decay scheme other than that of ^{67}Ga.

The most recent data indicate the appearance of at least two X-rays at energies below 10 keV, which may indicate the presence of ^{65}Zn and one (or more) of the long-lived cobalt isotopes ^{56}Co, ^{57}Co, and ^{58}Co.

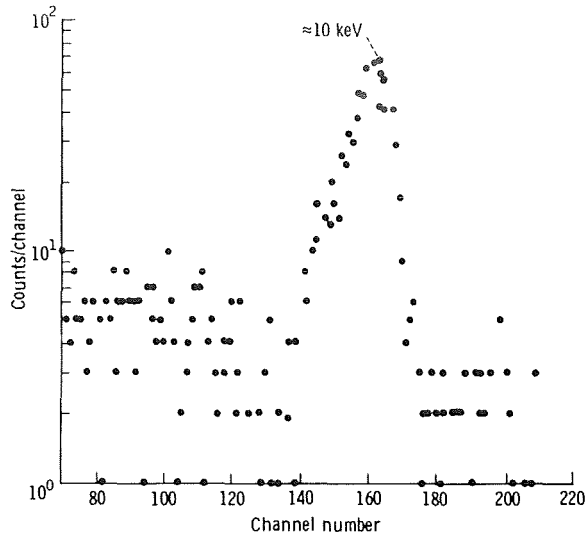

FIGURE 7-4.—Internal germanium spectrum showing doublet structure of 10-keV peak contributed by ^{67}Ga and ^{71}Ge (440-minute count time at 0.066 keV/channel).

Los Alamos Scientific Laboratory

The metal foil disks arrived at Los Alamos Scientific Laboratory 23 hours after splashdown. Analysis of the exposed foils was conducted as follows.

1. The scandium disk was nondestructively counted for ^{46}Sc to determine the thermal neutron-capture product.
2. Neptunium-239 (^{239}Np) was chemically separated from uranium and counted to determine the epithermal neutron-capture product.
3. Molybdenum-99 (^{99}Mo) was also chemically separated to measure the fission component.
4. The yttrium disk was dissolved and analyzed for ^{87}Y, an energetic neutron- and proton-induced reaction product, and for zirconium-89 (^{89}Zr), a low-energy-proton reaction product.
5. The titanium lid was chemically processed for scandium radionuclides produced by 1- to 20-MeV neutrons.
6. The stainless steel can was nondestructively analyzed for activities produced from iron.

The thermal neutron flux was 0.71 neutron/cm^2 • sec and the upper limit to the epithermal neutron flux was 0.003 neutron/cm^2 • sec. Table 7-I shows the average cross sections for reactions measured in the ASTP foils as well as the flux of particles above 1 MeV and above the approximate threshold energy for each reaction determined from the ASTP foil measurements.

Oak Ridge National Laboratory and JSC

Direct measurements of induced radionuclides were made by optically sealing a photomultiplier tube to the activated NaI(Tl) crystal. Pulse height spectra of the internal response were obtained beginning approximately 26 hours after splashdown. Such measurements are continuing to characterize the long-lived components. Indirect measurements of the induced radionuclides were made by using Ge(Li) detectors and a large anticoincidence-shielded NaI(Tl) spectrometer in the low-level facility at Oak Ridge National Laboratory (ref. 7-8).

Preliminary results from the low-level spectrometer system (4π system) yielded positive identification of iodine-124 (^{124}I), ^{126}I, and ^{24}Na in the NaI flight detector. Tentative identification of ^{22}Na was also made, but verification must await refinement of the spectral analysis. Semiquantitative disintegration-rate values are approximately one-third to one-half those found in the Apollo 17 experiment (ref. 7-6) for ^{124}I, ^{126}I, and ^{24}Na.

Gamma-gamma coincidence data on the germanium experiment from the 4π system indicated the presence of at least one short-lived positron-emitting radionuclide. Decay-curve analysis of the positron-positron coincidences yielded a half-life value of 29 hours. The early measurements also show apparent gamma-gamma coincidences of 0.5 with 0.8-MeV photon and of 0.8 with 1.4-MeV photon. The radionuclide most closely fitting these characteristics is arsenic-72 (^{72}As), a 26.0-hour positron emitter with a 0.834-MeV gamma ray in coincidence with its major positron branch. The positron-positron counting rate corrected to splashdown is approximately 2.7 counts/min with a corresponding system background of 0.1 count/min. At 41 days after splashdown, the positron-positron counting rate was only 50 percent above background. Detailed analysis of Ge(Li) spectra shows a tentative identification of ^{67}Ga, a 78-hour radionuclide that decays by electron capture.

TABLE 7-I.—Summary of Threshold Reactions[a]

Reaction	$\phi\sigma$, $m^2/cm^2 \cdot sec$ ($b/cm^2 \cdot sec$)		$E > 1$ *MeV*		*Threshold,* *MeV*	$E >$ *threshold*	
			$\langle\sigma\rangle$, m^2 (b)	ϕ, $cm^{-2}\ sec^{-1}$		$\langle\sigma\rangle$, m^2 (b)	ϕ, $cm^{-2}\ sec^{-1}$
^{238}U(n,f)	4.30×10^{-28}	(4.30)	1.48×10^{-28} (1.48)	2.9	1	1.48×10^{-28} (1.48)	2.9
Ti(n,x)^{47}Sc	.087	(.087)	.030 (.030)	2.9	2	.035 (.035)	2.5
Ti(n,x)^{46}Sc	≤.32	(≤.32)	.023 (.023)	≤36	5	.035 (.035)	≤24
^{27}Al(n,α)^{24}Na	≤15.5	(≤15.5)	.014 (.014)	≤1100	5	.023 (.023)	≤670
Ti(n,x)^{48}Sc	≤.23	(≤.23)	.0073 (.0073)	≤31	5	.0115 (.0115)	≤20
^{45}Sc(n,2n)^{44m}Sc	≤.22	(≤.22)	.018 (.018)	≤12	10	.035 (.035)	≤6
^{89}Y(n,2n)^{88}Y	≤1.65	(≤1.65)	.110 (.110)	≤15	10	.215 (.215)	≤8
^{89}Y(n,3n)^{87}Y	≤.41	(≤.41)	.058 (.058)	≤7.1	20	.133 (.133)	≤3.2

[a]Uncertainty in the fluxes is estimated to be approximately ± 50 percent. Symbols are defined as follows: E is energy; σ, cross section; ϕ, flux; n, neutron; f, fission; α, alpha particle; and x, X-ray.

DISCUSSION

The results of both internal and external monitoring of the NaI crystal showed an activation spectrum similar to that of the Apollo 17 crystal but at approximately one-third the intensity. Gallium-67 has been positively identified in the intrinsic germanium as well as in the external counting of the bulk germanium. Germanium-71 has been tentatively identified internally and ^{72}As, externally.

Orbital integrations of trapped proton fluxes give a mission-averaged contribution of 0.6 proton/cm^2 • sec for protons with energy greater than 100 MeV. This analysis also enables the mission-averaged cosmic ray flux to be estimated at 0.8 particle/cm^2 • sec. The quantities of all positively identified radionuclides are considerably in excess of predictions based on these fluxes.

These results clearly confirm that considerable fluxes of secondary neutrons are produced by the interaction of cosmic rays with heavy spacecraft, as was the case in Apollo 17 (ref. 7-6). A considerable flux of thermal neutrons was again shown by the ^{24}Na result and confirmed by the scandium activation. The very low epithermal neutron flux determined from the uranium foil disk is consistent with the low ^{128}I:^{24}N ratio found in the Apollo 17 crystal. The other species observed in the NaI crystal require an energetic neutron flux (10 to 100 MeV) of 2 neutrons/cm^2 • sec, consistent with the threshold detectors.

The reduction of activation between Apollo 17 and Apollo-Soyuz is approximately the same as the reduction in primary cosmic ray intensity due to geomagnetic shielding, showing that for this range the secondary neutron production is directly proportional to the primary cosmic ray intensity.

The production of secondary neutrons in heavy spacecraft and their efficient thermalization in Apollo modules again emphasize the necessity for developing codes to assess neutron production and transport, and demonstrate that gamma-ray spectrometers are best used far removed from heavy material.

The calculations used here are being improved by comparison with controlled monoenergetic irradiation experiments, which will also yield important information on the short half-life radionuclides missed by such return procedures.

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